

Dynamical Quantum Phase Transitions in presence of a spin bath

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(Dated: November 16, 2016)

We derive an effective time independent Hamiltonian for the transverse Ising model coupled to a spin bath, in the presence of a high frequency AC magnetic field. The spin blocking mechanism that removes the quantum phase transition can be suppressed by the AC field, allowing tunability of the quantum critical point. We calculate the phase diagram, including the nuclear spins, and apply the results to Quantum Ising systems with long-range dipolar interactions; the example of $LiHoF_4$ is treated in detail.

PACS numbers:

“Quantum phase transitions” (QPT) take place between bulk equilibrium phases in the zero-temperature ($T \rightarrow 0$) limit. Hertz [1] showed that finite- T thermodynamic and transport properties near the zero- T quantum critical point (QCP) should be determined solely by the nature of the QCP itself. Classic examples are the Quantum Ising system, and the paramagnetic/ferromagnetic (PM/FM) transition in strongly-correlated conductors. However in real experimental systems things are not so simple: in zero- T PM/FM transitions, disorder and 1st-order phase transitions often obscure the physics, and in solid-state Quantum Ising systems “environmental” spin bath modes [2] can suppress the QCP entirely [3, 4]. This is unfortunate, given the importance of Quantum Ising phenomenology in so many areas of physics. There currently exists no good theory of QPT in Ising systems in the presence of a spin bath; however, the external control of the spin bath decoherence for qubits has been studied from the perspective of Nuclear Magnetic Resonance, where sequences of pulses are used to manipulate the coupling of qubits to the environmental modes[5–9].

In this letter we address this problem by: (i) Enlarging the QPT scenario for Quantum Ising systems, by generalizing the theory to the case of a strong high-frequency AC field, and (ii) showing how in principle this allows the manipulation of the effective Ising Hamiltonian, enabling one to suppress the spin bath effects. The AC field creates a new effectively time-*independent* Hamiltonian for the system, inducing new interactions and suppressing others, thereby opening up a new class of QPTs for investigation. By varying the frequency, and intensity of the field, one also obtains a very rich zero- T phase diagram, with various new kinds of QCP.

Well-known solid-state examples of experimental Quantum Ising systems with long-range dipolar inter-spin interactions include the $LiHo_xY_{1-x}F_4$ rare earth system ([3, 4] and [10]–[17]), and the transition metal-based Fe_8 molecular spin system [20]. Recent experiments on 1-dimensional ion trap Quantum Ising chains (where spin bath effects may be entirely absent) have

also successfully varied the range of the interactions [21, 22]. These systems are all described at low energies by Hamiltonians with spins $\vec{\tau}_i$ truncated to the lowest Ising doublet, separated from the next level by a gap $\Lambda_o \gg |V_{i,j}|, |A|$, where $V_{i,j}$ and A are the strengths of the inter-spin and hyperfine couplings. We have:

$$H(t) = - \sum_{i=1}^N [\Delta_o + \Gamma(t)] \tau_j^x - \sum_{i < j} V_{i,j} \tau_i^z \tau_j^z + H_{\text{HF}} \quad (1)$$

The total effective field here is the sum of a constant Δ_o and a time-dependent $\Gamma(t)$; typically these are not real magnetic fields, but effective fields acting in the Hilbert space of the Ising doublet. We now assume that the frequency ω of the time-dependent field $\Gamma(t)$ falls in the range $\Lambda_o > \hbar\omega \gg |V_{i,j}|, |A|$, allowing us to describe the AC field driving effects on the quantum Ising system by a standard Magnus expansion [23–28], in inverse powers of ω . The Magnus expansion is a very powerful method to extract the stroboscopic time evolution of a time dependent system, when the frequency of the driving field is large. By means of a transformation to the interaction picture, ie., $H(t) \rightarrow \tilde{H}(t) = U_1 H(t) U_1^\dagger - i U_1 \dot{U}_1^\dagger$, where $U_1 = e^{i \int dt \sum_j \Gamma(t) \cdot \tau_j^x}$, one can also capture the renormalization of parameters produced by non-perturbative effects of the field. The Magnus expansion then approximates the time dependent Hamiltonian by a time averaged one given by:

$$\mathcal{H} = \tilde{H}_0 + \frac{1}{\omega} \sum_{n=1}^{\infty} \frac{1}{n} [\tilde{H}_n, \tilde{H}_{-n}] \quad (2)$$

where \tilde{H}_n is the n -th Fourier component of $\tilde{H}(t)$, and terms $\sim O(|A|/\omega^m, |V_{i,j}|/\omega^m)$ ($m \geq 2$) are assumed negligible at high frequency.

(i) Dynamical Quantum Phase Transition: As a warm up we first consider a ‘pure’ Quantum Ising QPT, with no spin bath. We apply a linear AC field $\Gamma(t) = \Gamma_x \cos(\omega t)$, and find that $[\tilde{H}_n, \tilde{H}_{-n}] = 0$ for all n , leaving

only the zeroth Fourier component \tilde{H}_0 in Eq. (2) (see Supplementary information for details), and thus a *time-independent* effective Hamiltonian:

$$\mathcal{H}^o = - \sum_i \Delta_o \tau_i^x - \sum_{i,j>i} \left[\tilde{V}_{i,j}^{zz} \tau_i^z \tau_j^z + \tilde{V}_{i,j}^{yy} \tau_i^y \tau_j^y \right] \quad (3)$$

where $\tilde{V}_{i,j}^{zz}(\alpha) = V_{i,j} [1 + \mathcal{J}_0(2\alpha)]/2$, $\tilde{V}_{i,j}^{yy}(\alpha) = V_{i,j} [1 - \mathcal{J}_0(2\alpha)]/2$, the dimensionless parameter $\alpha = \Gamma_x/\omega$ and $\mathcal{J}_m(\alpha)$ is an m -th order Bessel function; the superscript in \mathcal{H}^o indicates zero hyperfine couplings. Thus the periodic driving modifies the direction and strength of the interspin coupling tensor, and transforms the Ising model into an XY (strictly a YZ) model with anisotropy controlled by α (as previously shown by [23] in 1D). Note Eq.3 is valid in arbitrary dimension, but the dimensionality will play a role when calculating the different statistical averages, as we will discuss below.

The anisotropic XY model has Ising phase transitions when the transverse magnetic field $\Delta_o = \pm \tilde{V}_{i,j}^{\mu\mu}$, as well as anisotropic transitions for $\tilde{V}_{i,j}^{yy} = \tilde{V}_{i,j}^{zz}$, where the magnetization changes its orientation between M_y^o and M_z^o (the super-index indicate the absence of a spin bath in this section). To characterize the QPT we calculate the double-time Green's functions[29] using a $1/Z$ expansion to lowest order (Z is the coordination number), which coincides with the Random Phase Approximation[4, 30] (see Supplementary information for details). The $1/Z$ scaling implies that the results are expected to be more accurate in higher dimensional systems such as the 3D Ising model, which is considered here to describe $LiHo_xY_{1-x}F_4$. From the resulting Green's functions we derive the corresponding "self consistency equations" (SCE) for the magnetization:

$$M_\mu^o = \frac{B_\mu + M_\mu^o \tilde{V}_0^{\mu\mu}}{2\tilde{\omega}_S} \tanh\left(\frac{\beta\tilde{\omega}_S}{2}\right) \quad (4)$$

where M_μ^o ($\mu = x, y, z$) is the magnetization along the rotated μ -axis, $\tilde{\omega}_S = \sqrt{\sum_\mu \left(B_\mu + M_\mu^o \tilde{V}_0^{\mu\mu}\right)^2}$, the effective field is $B_\mu = (\Delta_o, 0, 0)$, and $\tilde{V}_0^{\mu\mu}$ is the $\mathbf{q} = 0$ Fourier component of the effective spin-spin interaction along μ . The inverse temperature β in Eq.4 corresponds to the phonon bath temperature that in general, would couple to the spin system. Although its definition is not generally possible in the presence of a driving field, we will discuss below how, under some circumstances, one can still make use of it.

Note that the formalism applied here is valid for arbitrary large spins (important to describe the 7/2 spin bath in the next section). The system's dimension is encoded in the coordination number Z , or equivalently in the zeroth Fourier component of the interaction potential V_0 . For the simplest case of nearest neighbor interaction, one finds that they are related by $V_0 = ZV$, however

in more general situations, as for the case of long-range dipolar interactions, this relationship fails and it is more convenient to just estimate V_0 by other means, keeping in mind that the system's dimension is somehow encoded in this value. It is also important to realize that, to lowest order in the $1/Z$ expansion, one neglects quantum correlations, which would also be affected by the dimensionality. Their contribution in general would modify the results, but their effect is increasingly small in higher dimensional systems (e.g., one should be specially careful if they are ignored in 1D, as the FM/PM transition is forbidden at finite temperature); therefore we would expect only small deviations for the case of $LiHo_xY_{1-x}F_4$.

From Eq.4 one can easily compare the driven and undriven case. For $\Gamma(t) = 0$ the pure 3D Ising model \mathcal{H}^o has the $T = 0$ solutions:

- For $\Delta_o < V_0^{zz}/2$:

$$M_x^o = \frac{\Delta_o}{V_0^{zz}}, \quad M_y^o = 0, \quad M_z^o = \pm \frac{\sqrt{(V_0^{zz})^2 - 4\Delta_o^2}}{2V_0^{zz}}. \quad (5)$$

- For $\Delta_o > V_0^{zz}/2$:

$$M_x^o = \frac{1}{2}, \quad M_{y,z}^o = 0. \quad (6)$$

The critical field is given by $B_c = V_0^{zz}/2$, and the finite- T solution gives a zero field Curie temperature $T_c = V_0^{zz}/4$. For the case of long-range dipolar interactions one can directly estimate the zeroth Fourier component V_0^{zz} by numerical means[4], or as we have done in our case, extract its value from experimental measurements[31]. The phase diagram is shown in Fig.1, where the dashed black line separates a spin ordered FM phase for $\Delta_o < B_c$ and a paramagnetic one otherwise.

If we add the AC field $\Gamma(t)$ the system is now described by Eq.3. The anisotropy factor $\tilde{V}_{j,l}^{zz} - \tilde{V}_{j,l}^{yy} = \mathcal{J}_0(2\alpha) V_{j,l}$ becomes a function of α and we recover the Ising model when $\mathcal{J}_0(2\alpha) = 0$. This means that anisotropic quantum phase transitions happen every time we cross $\mathcal{J}_0(2\alpha) = 0$, and the magnetization changes its direction between M_z^o and M_y^o for temperatures below a critical T_c . Ising transitions could also be induced when α is tuned, as the critical field B_c now oscillates between a maximum and a minimum value (red solid line in Fig.2). Therefore, if the DC field is within this window, we would also observe PM/FM transitions.

(ii) Nuclear Spin Bath effects: In Quantum Ising systems the spin bath effects are often dominated by a single species of nuclear spin I_r^μ at positions \mathbf{r}_r . Let us assume an effective hyperfine coupling

$$H_{\text{HF}} = \sum_{\mu, r, j} A_{r,j}^\mu I_r^\mu S_j^\mu \quad (7)$$

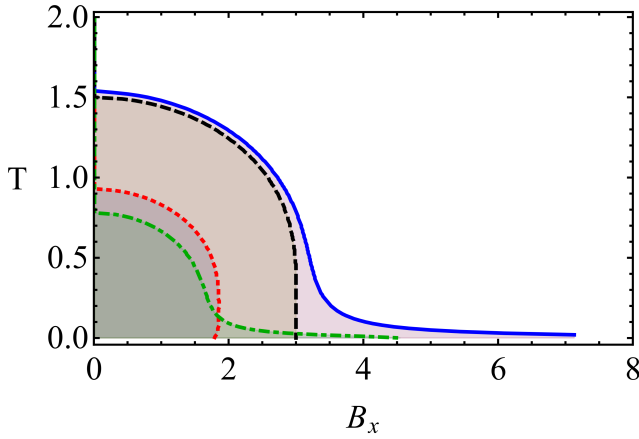


FIG. 1: Phase diagram of the AC driven transverse Ising model vs transverse field B_x and temperature. The dashed black line corresponds to the undriven system in absence of hyperfine coupling. The blue line corresponds to the undriven system coupled to the $I = 7/2$ spin bath. The red line corresponds to the AC driven system coupled to the spin bath for $\Gamma_x/\omega \sim 2.4$ (i.e., $\mathcal{J}_0(\alpha) = 0$). The green line corresponds to the case $2\Gamma_x/\omega \sim 2.4$, where the spin-spin interaction is symmetrical and resembles the Ising system again. For the plots we used: $V_0^{zz} = 6$ K and $A_0^{z,(\perp)} = 0.2$ (0.02) K. These values are chosen so that the phase diagram agrees with the experimental data in [31].

with principal axes along $\mu = x, y, z$ (the generalization to more complex forms is straightforward). Two spin bath mechanisms can then strongly affect Quantum Ising systems:

(i) *Transverse blocking mechanism*: In a quantum Ising system with hyperfine coupling, the electronic spin cannot simply flip between $|\uparrow\rangle$ and $|\downarrow\rangle$; it must carry the nuclear spin with it. However, transitions between $|\downarrow\uparrow\rangle$ and $|\uparrow\downarrow\rangle$ can then no longer be mediated by Δ_o , which do not flip the nuclear spin. Transverse hyperfine interactions could produce this flip, but in many real Quantum Ising systems, the effective longitudinal hyperfine coupling A_0^z is often much stronger than the transverse one (the large g factor anisotropy of any Ising system also forces a strong anisotropy in the Ising hyperfine coupling). The spin bath then strongly suppresses transverse electronic spin fluctuations [10], and the system reverts to classical Ising behavior until Δ_o is large enough to overcome A_0^z . This changes the phase diagram, shifting the critical point towards large values of Δ_0 (see Fig. 1, blue line), and it also radically alters the electronic spin dynamics. Many features of the resulting experimental behavior (such as the gapping of the electronic exciton mode in *LiHo*, even at the QCP [3]), are still not properly understood.

(ii) *Spin bath decoherence*: The spin bath causes decoherence in the electronic spin dynamics [2, 20]. Such decoherence blocks the use of Quantum Ising systems as

quantum information processors, for which they are otherwise ideally suited.

It would clearly be desirable to control the strength of both the inter-spin and the hyperfine couplings, and if possible, to suppress the hyperfine coupling completely. As we now see, this can be done in strong AC fields. We treat the hyperfine coupling $H_{\text{HF}} = \sum_{r,j,\mu} A_{r,j}^\mu I_r^\mu S_j^\mu$ in an AC field using the same maneuvers as above; H_{HF} is then renormalized to:

$$\mathcal{H}_{\text{HF}} = \sum_{\mu,j,r} \tilde{A}_{r,j}^\mu I_r^\mu S_j^\mu \quad (8)$$

where the effective coupling is $\tilde{A}_{r,j}^\mu = (A_{r,j}^x, A_{r,j}^y \mathcal{J}_0(\alpha), A_{r,j}^z \mathcal{J}_0(\alpha))$. Because the field couples to the electronic spins, but not to the nuclear spins (the nuclear Zeeman coupling $\ll |A_{r,j}^\mu|$), the renormalization of $A_{r,j}^\mu$ is different from that of $V_{i,j}$ (with factors $\mathcal{J}_0(\alpha)$ rather than $(1 \pm \mathcal{J}_0(2\alpha))/2$, and with α rather than 2α in the argument). Thus, by tuning the AC field amplitude, we can either (i) tune the inter-spin interaction $V_{i,j}$, to study of spin bath effects, or (ii) suppress the longitudinal hyperfine coupling A_z , to study the effects of $V_{i,j}$ in isolation.

To quantify all of this, we again derive self-consistent equations for the magnetization, now including the hyperfine coupling to the nuclear spin bath (to be specific this is done for the case $I = 7/2$, appropriate to the *LiHo* system). We then arrive at the pair of equations for the magnetization of the electronic system and nuclear bath (M_μ and m_μ , respectively):

$$M_\mu = \frac{B_\mu + M_\mu \tilde{V}_0^{\mu\mu} - m_\mu \tilde{A}_0^\mu}{2\tilde{\omega}_S} \tanh\left(\frac{\beta\tilde{\omega}_S}{2}\right) \quad (9)$$

$$m_\mu = -\frac{\tilde{A}_0^\mu M_\mu}{2\tilde{\omega}_B} \left[\tanh\left(\frac{\beta\tilde{\omega}_B}{2}\right) + 2 \tanh(\beta\tilde{\omega}_B) + 4 \tanh(2\beta\tilde{\omega}_B) \right] \quad (10)$$

where $\tilde{\omega}_S = \sqrt{\sum_\mu (B_\mu + M_\mu \tilde{V}_0^{\mu\mu} - m_\mu \tilde{A}_0^\mu)^2}$ is the system quasiparticle spectrum, and $\tilde{\omega}_B = \sqrt{\sum_\mu (M_\mu \tilde{A}_0^\mu)^2}$ is the spin bath quasiparticle spectrum. We see that if we set the field amplitude so that $\mathcal{J}_0(\alpha) = 0$, the longitudinal hyperfine coupling $\tilde{A}_0^{z,y}$ vanishes, and only the transverse part \tilde{A}_0^x remains. As for this value of α we have $\tilde{V}_0^{yy} > \tilde{V}_0^{zz}$, we find a ferromagnetic phase with magnetization along the y axis. Furthermore, as the hyperfine interaction only acts in the longitudinal direction, we find a renormalization of the critical field B_c to smaller values, but the QPT is still well defined, as it is driven by the transverse field Δ_0 (Fig. 1, dotted red).

Similarly, we can choose other values of α such as $\mathcal{J}_0(2\alpha) = 0$, where the asymmetry factor vanishes, and then the Ising model is recovered, or we can tune the sign of $\tilde{A}_0^{z,y}$, changing the ground state properties to a triplet

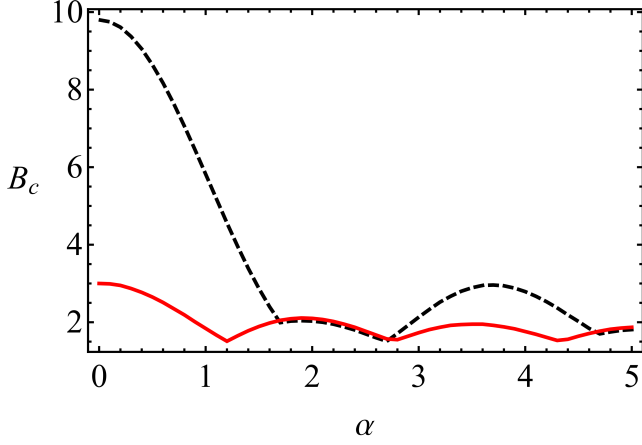


FIG. 2: The dashed black line shows the critical field B_c as a function of the ratio $\alpha = \Gamma_x/\omega$ for the AC driven Ising system coupled to a $7/2$ spin bath. The red solid line corresponds to the AC driven system in absence of the spin bath. At low amplitude, the spin bath contributes very strongly and drastically changes the critical field due to the blocking mechanism. As the amplitude increases, the effect of the bath is removed and only the transverse part A_0^x contributes with a small shift. There are some regions where B_c in presence of a bath is even lower than in the absence of hyperfine coupling ($\alpha \sim 1.7 - 2.7$). These are the regions where $\tilde{A}_0^{y,z}$ changes sign and overcomes A_0^x . The parameters are fixed according to the experimental ones for $LiHoF_4:V_0^{zz} = 6$ K, $A_0^z = 0.2$ K and $A_0^{x,y} = 0.02$ K.

state $\{|\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$. In Fig.2 we plot the critical field B_c for the AC driven Ising model coupled to the spin bath, as a function of the AC field parameter α , for $T = 0$.

This plot shows that for small α , the system behaves as in the undriven case, where the spin bath greatly affects the value of the critical field due to the blocking mechanism. As this blocking is produced by the difference between the transverse A_0^x and the longitudinal hyperfine coupling $A_0^{y,z}$, as the later is renormalized by the AC field we observe that when α is increased, the system approaches the isolated system behavior. Therefore, it would be possible to experimentally analyze the opposite regimes of ideal Ising QPT in absence of a spin bath and in presence of a spin bath by just tuning the external AC field.

(iii) Experiments and Conclusions: We have seen how one can obtain a static effective Hamiltonian for the AC driven transverse Ising model in presence of a spin bath, in which the inter-spin and the hyperfine interactions are renormalized and depend on the AC field intensity and frequency. This shows that one can study the Ising QPT in the presence of a spin bath, and even remove its effect. Regarding the effect of higher order frequency corrections, we have calculated the next order leading term

$\frac{1}{2\omega^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \left(\left[[\tilde{H}_n, \tilde{H}_0], \tilde{H}_{-n} \right] + h.c. \right)$, and although the effective Hamiltonian contains now up to four-body interactions, they are all weighted by Bessel functions and a factor ω^{-2} , which in general give corrections one or two orders of magnitude smaller than \tilde{H}_0 . Importantly, we find that the transverse blocking mechanism, produced due to the initially large anisotropy between A_0^z and $A_0^{x,y}$ is not restored by the second order corrections, and the renormalized critical point should not be greatly affected. Nevertheless one should be careful with the growth of higher frequency corrections for large times, which would restrict the maximum duration of the experiments.

We now discuss the experimental setups. The classic QPT magnetic insulator $LiHo_xY_{1-x}F_4$, with spin $S = 8$ magnetic ions, perhaps the canonical Quantum Ising system, displays quantum annealing [11] and a quantum spin glass phase [13], as well as quantum critical behavior [14]. However, the strong coupling to the nuclear spin bath disrupts completely the expected quantum critical behavior around the QCP [3, 10] (and leads to various other dynamic and thermodynamic effects [15–17]). For nearest neighbor spins, $|V_{i,j}| \rightarrow V \sim 1.2x$, in K units, and the hyperfine level splitting $|A| \sim 0.22 K$ (with spin $I = 7/2$). In a high frequency AC field ($\Lambda_o \gg \hbar\omega \gg |V_{i,j}|, |A_{r,j}^{\mu}|$, ie, for $\omega \sim 30 - 200$ GHz), we may then directly apply the theory given here. The results are shown in Figs. 1 and 2), and these constitute predictions for this system.

In the Fe_8 system the hyperfine couplings are much smaller, and can be varied by isotopic substitution[18]. Because there is a whole spectrum of these couplings, one cannot suppress them all simultaneously - but one can select out particular groups of nuclear spins for suppression, and the effective couplings as a function of applied field are well understood [19, 20]. What is interesting here is the possibility of controlling the longitudinal dipolar coupling between molecules, allowing one to look at single molecule dynamics.

In ion trap spin chains we can typically discount spin bath effects. What is interesting here is the possibility of varying the range of the inter-spin interactions, as well as their strength, and observing the spin dynamics in real time for both short- and long-range interaction forms [22]; calculations are underway to give quantitative predictions. In these systems one can also introduce transverse inter-spin interactions - this makes the eventual phase diagram very rich indeed.

In all three systems experimental testing of the results herein should be easily possible - quantitative comparison will require numerical work, and we emphasize that in real experiments one will need to take account of the following factors:

(a) Demagnetization fields, which are in general inhomogeneous in real systems, will need to be evaluated

numerically (compare [20]); experiments with “whisker”-shaped samples (for solid-state systems) would be useful.

(b) In solid-state systems, heating of the sample needs to be avoided. A pulsed field experiment would be useful, with the pulses short enough so that the spin-phonon couplings have no time to heat the phonon bath. The initialization protocol should be engineered so as to reach the desired Floquet steady state, which can be done us-

ing adiabatic launching. Furthermore, interactions with a phonon bath could be used to stabilize the steady state[32]. Finally, the field amplitude of Γ_x has to be of the order of the frequency.

This work has been supported by NSERC of Canada, and by PITP. We acknowledge helpful discussions with G. Aeppli on the experimental constraints.

Appendix A: calculation of the effective hamiltonian

Here we first discuss the relationship between the high-energy and low-energy spin Hamiltonians, and then go on to show how one derives the effective Hamiltonian for the Quantum Ising system in a strong high-frequency AC field, including details of the corrections to the lowest order Magnus expansion term used in the main text.

(i) TRUNCATION to the LOW-ENERGY ISING HAMILTONIAN: In discussing Quantum Ising systems we will mostly work with a low-energy description in which the electronic levels for each magnetic ion have been truncated to a lowest-energy doublet. This will be described by the effective Hamiltonian (eqtn. (1) of the main text):

$$H_{eff}(t) = - \sum_i [\Delta_o + \Gamma(t)] \tau_i^x - \frac{1}{2} \sum_{i,j \neq i} V_{i,j} \tau_i^z \tau_j^z + H_{HF}^{eff} \quad (A1)$$

where $\hat{\tau}_j^x$ acts in the 2-dimensional subspace of the lowest doublet on site i , and the transverse fields Δ_o and $\Gamma(t)$ are not the actual fields applied to the system, but the effective fields acting in the same low-energy Hilbert space. The same applies to $V_{i,j}$, the effective interaction between the Pauli spins on sites i and j , and to the hyperfine interaction H_{HF} , which in this low-energy Hilbert space has the general form

$$H_{HF}^{eff} = \sum_{j,k} A_{j,k}^{\mu\nu} \tau_j^\mu I_k^\nu \quad (A2)$$

where $A_{j,k}^{\mu\nu}$ is the effective low-energy anisotropic hyperfine coupling between the electronic spin doublet at site j and a nuclear spin at site k .

The low-energy effective Hamiltonian is truncated from a spin Hamiltonian of form

$$H(t) = - \sum_i H_o(\mathbf{S}_j) + [B_x + H_x(t)] S_i^x - \frac{1}{2} \sum_{i,j \neq i} U_{i,j} S_i^z S_j^z + H_{HF} \quad (A3)$$

where $H_o(\mathbf{S})$, the local “high-energy” ionic spin Hamiltonian, acts on spins $\{\mathbf{S}_j\}$; the Hilbert space for each magnetic ion now has dimension $2S + 1$. The high-energy hyperfine coupling takes the form

$$H_{HF} = \sum_{\mu,\nu} \sum_{j,k} \Lambda_{j,k}^{\mu\nu} S_j^\mu I_k^\nu \quad (A4)$$

where we use a slightly unconventional notation in which $\Lambda_{jk}^{\mu\nu}$ denotes the “bare” hyperfine coupling between the full spin \mathbf{S}_j and the nuclear spin \mathbf{I}_k , and drop the nuclear quadrupolar couplings, which are negligible for the quantum Ising systems examined so far.

While the low-energy form (Eq.A1) is generic to all of the Quantum Ising systems so far investigated, the high-energy form (Eq.A3) varies widely from one physical system to another, depending on the magnitude of the \mathbf{S}_j , the lattice symmetry, the strength of the spin-orbit, crystal field, and hyperfine fields, and so on. Thus the details of the truncation, of the dependence of the low-energy fields Δ_o and $\Gamma(t)$ on external field, and of the magnitude and anisotropy of the low-energy $A_{j,k}^{\mu\nu}$ and $V_{i,j}$, depend very much on which system we are looking at. We consider here the 3 cases mentioned in the main paper:

(a) *LiHo_xY_{1-x}F₄*: In this case, the high-energy Hamiltonian involves ionic spins with $S = 8$ and a local spin Hamiltonian

$$H_o(\mathbf{S}) = \sum_{k=2,4,6} B_k^0 \hat{O}_k^0 + \sum_{k=4,6} B_k^4(C) \hat{O}_k^4(C) + B_6^4(S) \hat{O}_6^4(S) \quad (\text{A5})$$

written in terms of the standard Stevens operators \hat{O}_k^q (see, eg., Jensen and MacKintosh [33]); the best values of the parameters B_k^q are given by Ronnow et al [34]. This "high-energy" form is valid up to energy scales $\sim 10^3 K$. The truncation of the high-energy Hamiltonian of eqtns. (A3)-(A5) down to the low-energy form in eqtns. (A1) and (A2) has been thoroughly discussed in the literature [34-36]. The low-energy form can be used for energies smaller than the gap of $\sim 11.3 K$ which exists between the low-energy spin doublet and a 3rd intermediate state, through which virtual transitions allow a coupling between the 2 lowest Ising states $|\uparrow\rangle$ and $|\downarrow\rangle$ on each site. The transition matrix element $\Delta_o(B_x)$ between these states is a highly non-linear function of B_x , obtainable by exact diagonalization [36]. The hyperfine interactions are large: experiment shows that $\Lambda_{j,k}^{\mu\nu} \sim \Lambda_o \sim 0.039 K$ for the bare on-site *Ho* hyperfine coupling, for which $I = 7/2$, with considerably smaller values for the hyperfine couplings to the four *F* nuclear spins. This then gives a splitting between adjacent hyperfine levels of $\sim 0.22 K$, and a total spread of energy over the 8 hyperfine levels of $\sim 1.5 K$. Thus the hyperfine energy scale competes very well with the dipolar coupling $U_{i,j}$ between nearest neighbor *Ho* ions, even for the pure *LiHo* system, where the energy difference between $|\uparrow\uparrow\rangle$ and $|\uparrow\downarrow\rangle$ configurations coming from the dipolar interactions is also $\sim 1.5 K$; for *Y*-doped *LiHo_xY_{1-x}F₄*, this nearest neighbor dipole coupling is reduced by a factor $\sim O(x)$, and the hyperfine coupling then dominates.

(b) **The *Fe₈* molecular spin system**: In this case the high-energy Hamiltonian has spin $S = 10$ (coming from a core of 8 *Fe* ions), and a local spin Hamiltonian which is well approximated by

$$H_o(\mathbf{S}) = -D_o \hat{S}_x^2 + E_o \hat{S}_y^2 - K_4 (\hat{S}_+^4 + \hat{S}_-^4) \quad (\text{A6})$$

where the values of D_o, E_o , and K_4 (all positive) were measured some time ago [37]; this form can be used up to energy scales $\sim 50 K$. Each *Fe₈* molecule has up to 213 different nuclear spins, depending on which of the various *Fe, Br, N, O*, and *H* isotopes in the molecule are being used; each of the hyperfine couplings for this system has been calculated, but they are typically very small (the proton couplings range from $\sim 3 mK$ down to well below $1 mK$, except for the odd outlier, with similar values for the other non-metallic nuclear species; the coupling to isotopically substituted ^{57}Fe nuclei is $\sim 4 mK$). Thus the intermolecular dipolar coupling, of a similar magnitude to that for *LiHo*, is much larger. The truncation to the low-energy doublet form can again be done numerically [19], and is valid for energies smaller than the gap of $\sim 5 K$ to the next highest states. Again the dependence of Δ_o and $\Gamma(t)$ on a transverse field \mathbf{H}_\perp is very non-linear, and also varies enormously with the angle of the field in the easy $\hat{x}\hat{y}$ -plane - for \mathbf{H}_\perp oriented along the easy \hat{x} -axis one sees very strong oscillations of Δ_o as a function of H_x .

(c) **Ionic spin chains**: In this case one deals with systems where we can, to a good approximation, begin by ignoring the coupling to a spin bath. The Hamiltonian in experiments [39-41] can be more general than the standard Quantum Ising system; the low-energy Hamiltonian is

$$\mathcal{H}_{eff} = \Delta_o \sum_j \tau_j^x + \sum_{i \neq j} J_{i,j}^{zz} \tau_i^z \tau_j^z + \mathcal{H}_J^\perp \quad (\text{A7})$$

where the extra term is just an *XY* form $\mathcal{H}_J^\perp = \sum_{i \neq j} J_{i,j}^\perp (\tau_i^+ \tau_j^- + \tau_i^- \tau_j^+)$. The parameters Δ_o and $J_{i,j}^{\alpha\alpha}$ are again effective parameters, related to the original applied fields in ways described in detail in refs. [39]-[41]. In different experiments with different ions one can vary these parameters over a rather wide range; typical values are $10^{-9} K < J_o < 10^{-7} K$, and $10^{-2} < \Delta_o/J_o < 5$, where J_o is a typical nearest neighbor value for either $J_{i,j}^{zz}$ or $J_{i,j}^\perp$. The interactions typically take a power law form as a function of the distance $r_{ij} = a_o|i-j|$, where a_o is the lattice spacing (typical 2-3 μm), ie., $|J_{i,j}^{\alpha\alpha}| \sim J_o|i-j|^{-p}$, where in principle one can vary p between 1 - 3. Provided Δ_o is not too large, the coupling to phonons can be adequately suppressed.

The results in what follows will apply only to the case where we ignore the "easy-plane" or *XY*-coupling terms in (A7); the phase diagram with these terms added becomes very rich and requires a separate study.

In what follows we will work exclusively with the low-energy effective Hamiltonian given in (A1) above, because our topic is the behavior of a Quantum Ising system in a high-frequency AC field. Thus we will leave the behavior of the parameters $\Delta_o, \Gamma(t), A_{jk}^{\mu\nu}$, and V_{ij} , as functions of the real applied fields, in the form of undetermined variables in this effective Hamiltonian, to be determined in practice by a combination of numerical calculation and experiment on whichever system one is dealing with.

QUASI-STATIC HAMILTONIAN in a STRONG HIGH-FREQUENCY AC FIELD: When the system is in the presence of a high frequency transverse field $H_x(t)$ with frequency ω , the stroboscopic time evolution can be approximated by an effective time independent evolution operator, obtained from a Magnus expansion. Corrections to this expression, due to finite frequency effects and to the initialization procedure can be important, and they must be analyzed carefully. We assume that the system frequency is such that $|A_{i,j}^{\mu\mu}|/\omega$ and $|V_{i,j}|/\omega$ are small. The Magnus expansion is then a expansion in powers of these parameters. In the main text, we have given expressions in which one drops corrections $\sim O(\omega^{-2})$ in the Magnus expansion, keeping only the lowest terms. In the discussion immediately below, we derive the form of these lowest terms (ie., terms $\sim O(1/\omega)$). At the end of this sub-section, we derive the next corrections, and show that for the present problem these can indeed be neglected.

In discussing a Magnus expansion, it is important to specify the "initialization protocol", ie., the way in which the AC applied field is ramped up at the beginning. One option often used is to use an "adiabatic launching protocol" [28], which consists in reaching the final non-equilibrium steady state by keeping the system in the same quasienergy state. Heating can be a problem here, specially due to the spin-phonon couplings in the system; however, these couplings are typically rather weak for the Quantum Ising systems being currently studied. This means that it will take the phonon bath some time to react to the rapid oscillations of the electronic spins. Thus the use of pulsed fields is more appropriate, with widely spaced pulses, as the system then has time for energy relaxation between pulses. It would also be helpful to have a phonon bath for which the density of states for undesired Floquet transitions, which could drive the system out of the steady state, is small; this would stabilize the Floquet phase described in this work [32].

The time evolution operator over a single period of the AC field can be written using a standard Magnus expansion in the general form:

$$U(\tau + T, \tau) = e^{iK(\tau)} e^{-i\mathcal{H}T} e^{-iK(\tau)} \quad (\text{A8})$$

where the unitary "kick operators" $K(\tau)$ contain all information about the starting time τ , $\mathcal{H} = \tilde{H}_0 + \frac{1}{\omega} \sum_n \frac{1}{n} [\tilde{H}_n, \tilde{H}_{-n}] + \mathcal{O}(\omega^2)$ and the sub-index n corresponds to the n -th Fourier mode of the time dependent Hamiltonian $\tilde{H}(t)$, which is specified below. As the spectrum is invariant under unitary transformations in Eq.A8, the effect of the τ dependent terms can be ignored, and neglecting $O(1/\omega^2)$ corrections, the effective Hamiltonian is given by the time averaged one \tilde{H}_0 and the finite frequency corrections $\frac{1}{\omega} \sum_n \frac{1}{n} [\tilde{H}_n, \tilde{H}_{-n}]$. To quantify the renormalization effects produced by a large amplitude of the AC field, we must first use a transformation to the interaction picture:

$$\tilde{H}(t) = U_1 H(t) U_1^\dagger - i U_1 \dot{U}_1^\dagger = - \sum_i B_x \tilde{\tau}_i^x - \frac{1}{2} \sum_{i,j \neq i} V_{i,j} \tilde{\tau}_i^z \tilde{\tau}_j^z + \sum_{\mu,r,j} A_{r,j}^\mu I_r^\mu \tilde{\tau}_j^\mu \quad (\text{A9})$$

where $U_1 = \exp \left[i \int \sum_j \mathbf{H}(t) \cdot \boldsymbol{\tau}_j \right] dt$, with $\mu = x, y, z$ and $\tilde{\tau}_j^\mu \equiv U_1 \tau_j^\mu U_1^\dagger$. For the calculation of the Magnus expansion we need the Fourier coefficients of the time dependent Hamiltonian. They are given by:

$$\tilde{H}_0 = - \sum_i B_x \tau_i^x - \sum_{i,j \neq i} \frac{V_{i,j}}{4} \{ [1 + \mathcal{J}_0(2\alpha)] \tau_i^z \tau_j^z + [1 - \mathcal{J}_0(2\alpha)] \tau_i^y \tau_j^y \} + \sum_{\mu,r,j} \tilde{A}_{r,j}^\mu I_r^\mu \tau_j^\mu \quad (\text{A10})$$

$$\tilde{H}_{\pm(2n+1)} = \pm i \sum_{i,j \neq i} \frac{V_{i,j} \mathcal{J}_{2n+1}(2\alpha)}{4} (\tau_i^y \tau_j^z + \tau_i^z \tau_j^y) \pm i \mathcal{J}_{2n+1}(\alpha) \sum_{r,j} (A_{r,j}^y I_r^y \tau_j^z - A_{r,j}^z I_r^z \tau_j^y) \quad (\text{A11})$$

$$\tilde{H}_{\pm 2(n+1)} = - \sum_{i,j \neq i} \frac{V_{i,j} \mathcal{J}_{2n+2}(2\alpha)}{4} (\tau_i^z \tau_j^z - S_i^y \tau_j^y) + \mathcal{J}_{2(n+1)}(\alpha) \sum_{r,j} (A_{r,j}^y I_r^y \tau_j^y + A_{r,j}^z I_r^z \tau_j^z) \quad (\text{A12})$$

where $\alpha \equiv H_x/\omega$, $\mathcal{J}_m(\alpha)$ is an m -th order Bessel function, and where the renormalized hyperfine couplings are

$$\tilde{A}_{r,j}^x = A_{r,j}^x; \quad \tilde{A}_{r,j}^y = A_{r,j}^y \mathcal{J}_0(\alpha); \quad \tilde{A}_{r,j}^z = A_{r,j}^z \mathcal{J}_0(\alpha). \quad (\text{A13})$$

From these expression we see that $[\tilde{H}_n, \tilde{H}_{-n}] = 0$, and to order $1/\omega$, we only need to use \tilde{H}_0 in the expansion. Thus, to order $1/\omega$, the effective time-independent Hamiltonian becomes:

$$\mathcal{H} = - \sum_i B_x \tau_i^x - \frac{1}{2} \sum_{i,j \neq i}^N [\tilde{V}_{ij}^{zz} \tau_i^z \tau_j^z + \tilde{V}_{ij}^{yy} \tau_i^y \tau_j^y] + \sum_{\mu,r,j} \tilde{A}_{r,j}^\mu I_r^\mu \tau_j^\mu \quad (\text{A14})$$

in which the renormalized dipolar couplings are:

$$\tilde{V}_{i,j}^{zz}(\alpha) = V_{i,j} [1 + \mathcal{J}_0(2\alpha)]/2 \quad (\text{A15})$$

$$\tilde{V}_{i,j}^{yy}(\alpha) = V_{i,j} [1 - \mathcal{J}_0(2\alpha)]/2 \quad (\text{A16})$$

We see that under the effect of the AC field the model becomes an effective anisotropic "XY" (actually, YZ) spin system in a transverse field. Thus one effect of the AC field is to modify the very strong dominance of the "zz" coupling in the effective dipolar interaction between Ising spins. We note also that the argument of the Bessel functions in the renormalized hyperfine couplings is half that involved in the renormalized inter-spin couplings.

One may worry that the terms to order $1/\omega^2$ in the expansion may change the form of the effective Hamiltonian in some important way, and/or that the corrections are not small. To see that this is not the case, as an example we have calculated the next order leading term. The result is a rather long expression, so we just discuss its general properties. The next term in the frequency expansion corresponds to:

$$\tilde{H}_2 = \frac{1}{2\omega^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \left(\left[[\tilde{H}_n, \tilde{H}_0], \tilde{H}_{-n} \right] + H.c. \right) \quad (\text{A17})$$

Its calculation displays 4-body interactions between the electronic spins (of "zx", "zy", "xy", and "yy" form, but no "zz" terms), along with 3-body terms $\propto B_x$, plus 4- and 3-body terms involving both electronic and bath spins. However they are all weighted by Bessel functions and a by factor ω^{-2} . This gives corrections much smaller than \tilde{H}_0 in the high frequency regime, as we can see by putting some numbers in. Thus, eg., if we assume that $\omega = 10|V|$, where V is a typical size for the nearest neighbor coupling $V_{i,j}$ between electronic spins, the second order correction is nearly two orders of magnitude smaller than \tilde{H}_0 .

In addition to this, as discussed in the main text, the transverse blocking mechanism is produced essentially by the initially large ratio between A_0^z and $A_0^{x,y}$ (with $A_0^z \gg A_0^{x,y}$). As we have just seen above (and in the main text), this large hyperfine "zz" anisotropy is removed by \tilde{H}_0 ; we therefore expect that the small corrections produced by the $O(\omega^{-2})$ terms will not change the character or position of the renormalized critical point derived below.

Appendix B: Green's function calculation

In what follows we first set up the general framework for deriving equations of motion for the appropriate Green's functions, and we then show how this works out in practice for specific cases.

(i) General Expressions for Correlators: A more general form of the Hamiltonian discussed in this paper is:

$$H = - \sum_{\mu,j} B_{\mu} S_j^{\mu} - \frac{1}{2} \sum_{\mu} \sum_{j,l \neq j} V_{j,l}^{\mu} S_j^{\mu} S_l^{\mu} + \sum_{\mu,r,j} A_{r,j}^{\mu} I_r^{\mu} S_j^{\mu} \quad (\text{B1})$$

where $\alpha, \beta = x, y, z$, and S_n^{α} operates on an arbitrary spin of spin S (not just $S = 1/2$) at site n ; the nuclear spin I_r^{μ} also takes arbitrary value. We are interested in the Green's function for the calculation of the magnetization, defined by:

$$G_{n,m}^{\alpha,\beta}(t, t') = -i \langle \langle S_n^{\alpha}(t); S_m^{\beta}(t') \rangle \rangle \quad (\text{B2})$$

where $\langle \langle \dots \rangle \rangle$ corresponds to the statistical average with respect to the thermal density matrix $\rho = e^{-\beta H}$, and the semi-colon indicates that we can consider the time ordered, retarded or advanced Green's functions (they all have the same equation of motion). The corresponding equation of motion for the electronic spins is given by:

$$\begin{aligned} \omega G_{n,m}^{\alpha,\beta}(\omega) &= \frac{1}{2\pi} \langle \{ S_n^{\alpha}, S_m^{\beta} \} \rangle + i \sum_{\mu} \epsilon_{\mu\alpha\delta} B_{\mu} G_{n,m}^{\delta,\beta}(\omega) \\ &+ i \sum_{\mu} \epsilon_{\mu\alpha\delta} \sum_{j \neq n} V_{n,j}^{\mu} G_{jn,m}^{\mu\delta,\beta}(\omega) - i \sum_{\mu,r} \epsilon_{\mu\alpha\delta} A_{r,n}^{\mu} K_{rn,m}^{\mu\delta,\beta}(\omega) \end{aligned} \quad (\text{B3})$$

where we have defined:

$$G_{jn,m}^{\mu\delta,\beta}(t, t') = -i \langle \langle S_j^{\mu}(t) S_n^{\delta}(t); S_m^{\beta}(t') \rangle \rangle, \quad K_{rn,m}^{\mu\delta,\beta}(t, t') = -i \langle \langle I_r^{\mu}(t) S_n^{\delta}(t); S_m^{\beta}(t') \rangle \rangle \quad (\text{B4})$$

The expression above is valid for arbitrary spin values. Note that we have used anti-commutation relationships for the definition of the Green's functions, as it is more convenient for the underlying pole structure that we will encounter later on. In what follows we adapt the spin operator decoupling methods discussed by, eg., Wang et al. [44], for lattice electronic spins, to the more general case of a set of lattice spins coupled to nuclear spins.

We decouple the higher Green functions in the equation of motion neglecting correlations between different sites. This approximation can be understood from the perspective of a $1/Z$ expansion, being Z the coordination number of the system. It is known that to lowest order, ie neglecting quantum correlations, the $1/Z$ expansion agrees with the Random Phase Approximation (RPA) and that for higher dimensional systems such as the 3D Ising model considered for *LiHoF*, it should provide reasonable good results[34]. Once we applied this decoupling scheme we find:

$$\omega g_{n,n}^{\alpha,\beta} = \frac{\chi_{\alpha,\beta}}{2\pi} + i \sum_{\mu} \epsilon_{\mu\alpha\delta} \left(B_{\mu} + \sum_{j \neq n} V_{n,j}^{\mu} \langle S_j^{\mu} \rangle - \sum_r A_{r,n}^{\mu} \langle I_r^{\mu} \rangle \right) g_{n,n}^{\delta,\beta} \quad (\text{B5})$$

where $\chi_{\alpha\beta} = \langle \{S_n^{\alpha}, S_n^{\beta}\} \rangle$ are the spin anti-commutators (we suppress the site index n) and $g_{n,n}^{\alpha,\beta}$ the Green functions in the RPA approximation. The calculation of the Green's functions is fairly straightforward [44]; we rewrite the system of equations as:

$$(\mathbf{1}\omega - \mathbf{H}) \mathbf{g}(\omega) = \mathbf{F}, \quad \mathbf{g}(\omega) = \begin{pmatrix} g_{n,n}^{x,\beta}(\omega) \\ g_{n,n}^{y,\beta}(\omega) \\ g_{n,n}^{z,\beta}(\omega) \end{pmatrix} \quad (\text{B6})$$

where

$$\mathbf{F} = \frac{1}{2\pi} \begin{pmatrix} \chi_{x,\beta} \\ \chi_{y,\beta} \\ \chi_{z,\beta} \end{pmatrix}, \quad \mathbf{H} = \begin{pmatrix} 0 & iH_z & -iH_y \\ -iH_z & 0 & iH_x \\ iH_y & -iH_x & 0 \end{pmatrix}, \quad (\text{B7})$$

and H_{α} are the components of an effective field defined as

$$H_{\alpha} = B_{\alpha} + M_{\alpha} V_0^{\alpha} - m_{\alpha} A_{\alpha}, \quad (\text{B8})$$

M_{α} is the electronic spin magnetization, and m_{α} the nuclear spin bath magnetization. The \mathbf{H} matrix can be diagonalized and has eigenvalues $\omega = \{0, \pm \sqrt{\sum_{\alpha} H_{\alpha}^2}\}$.

The Green functions can then be obtained as:

$$g_{n,n}^{\alpha,\beta} = \sum_{\lambda=1}^3 \sum_{\tau=1}^3 \frac{U_{\alpha\tau} U_{\tau\lambda}^{-1}}{\omega - \omega_{\tau}} F^{\lambda,\beta} = \sum_{\lambda=1}^3 R^{\alpha,\lambda} F^{\lambda,\beta} \quad (\text{B9})$$

where U is the matrix that diagonalizes \mathbf{H} . From this expression we calculate the statistical averages straightforwardly using:

$$\langle S_n^{\beta} S_n^{\alpha} \rangle = i \int \frac{g_{n,n}^{\alpha,\beta}(\omega + i\epsilon) - g_{n,n}^{\alpha,\beta}(\omega - i\epsilon)}{e^{\beta\omega} + 1} d\omega \quad (\text{B10})$$

$$\langle S_n^{\beta} S_n^{\alpha} \rangle = \sum_{\lambda=1}^3 \sum_{\tau=1}^3 \frac{U_{\alpha\tau} U_{\tau\lambda}^{-1}}{e^{\beta\omega_{\tau}} + 1} \tilde{F}^{\lambda,\beta} \quad (\text{B11})$$

where

$$\tilde{F}^{\lambda\beta} = 2\pi F^{\lambda\beta} = \chi^{\lambda\beta} \quad (\text{B12})$$

In order to simplify the expression further, we can use the relation between commutators and anti-commutators:

$$\langle \{S_n^{\alpha}, S_n^{\beta}\} \rangle = \langle [S_n^{\alpha}, S_n^{\beta}] \rangle + 2\langle S_n^{\beta} S_n^{\alpha} \rangle \quad (\text{B13})$$

Writing $\Gamma = \sum_{\tau=1}^3 \frac{U_{\alpha\tau} U_{\tau\lambda}^{-1}}{e^{\beta\omega_{\tau}} + 1}$, the matrix equation for the statistical averages then becomes

$$(\mathbf{1} - 2\Gamma) \begin{pmatrix} \langle S_n^{\beta} S_n^x \rangle \\ \langle S_n^{\beta} S_n^y \rangle \\ \langle S_n^{\beta} S_n^z \rangle \end{pmatrix} = \Gamma \begin{pmatrix} \langle [S_n^{\alpha}, S_n^{\beta}] \rangle \\ \langle [S_n^{\alpha}, S_n^{\beta}] \rangle \\ \langle [S_n^{\alpha}, S_n^{\beta}] \rangle \end{pmatrix} \quad (\text{B14})$$

We now define everything in terms of a dimensionless "eigenvalue normalized" effective field $h_\alpha = H_\alpha / \sqrt{\sum_\alpha H_\alpha^2}$, and a renormalized frequency $\tilde{\omega} = \omega / \sqrt{\sum_\alpha H_\alpha^2}$. The explicit calculation of the matrix equation then results in:

$$2 \begin{pmatrix} 0 & ih_z & -ih_y \\ -ih_z & 0 & ih_x \\ ih_y & -ih_x & 0 \end{pmatrix} \begin{pmatrix} \langle S_n^\beta S_n^x \rangle \\ \langle S_n^\beta S_n^y \rangle \\ \langle S_n^\beta S_n^z \rangle \end{pmatrix} = \begin{pmatrix} \coth\left(\frac{\beta\tilde{\omega}}{2}\right) & -ih_z & ih_y \\ ih_z & \coth\left(\frac{\beta\tilde{\omega}}{2}\right) & -ih_x \\ -ih_y & ih_x & \coth\left(\frac{\beta\tilde{\omega}}{2}\right) \end{pmatrix} \begin{pmatrix} i\epsilon_{x\beta\delta} \langle S_n^\delta \rangle \\ i\epsilon_{y\beta\delta} \langle S_n^\delta \rangle \\ i\epsilon_{z\beta\delta} \langle S_n^\delta \rangle \end{pmatrix} \quad (\text{B15})$$

In order to solve these coupled equations one must realize that they are not independent, as the determinant of $\mathbf{I} - 2\mathbf{\Gamma}$ vanishes. The first row multiplied by h_x , plus the second row times h_y , plus the third row times h_z gives zero. The same condition on the right reads:

$$\epsilon_{x\beta\delta} \langle S_n^\delta \rangle h_x + \epsilon_{y\beta\delta} \langle S_n^\delta \rangle h_y + \epsilon_{z\beta\delta} \langle S_n^\delta \rangle h_z = 0 \quad (\text{B16})$$

If we now choose $\beta = x, y, z$ we find, respectively:

$$\begin{aligned} \langle S_n^y \rangle h_z &= \langle S_n^z \rangle h_y \\ \langle S_n^z \rangle h_x &= \langle S_n^x \rangle h_z \\ \langle S_n^x \rangle h_y &= \langle S_n^y \rangle h_x \end{aligned}$$

which are the so called regularity conditions. They imply that we only need to know one of the components of the magnetization in order to calculate the other components.

Actually one can solve a somewhat more general system of equations, again for arbitrary spin. Consider an arbitrary polynomial function of spin operators of form

$$P(\{S_n^\alpha\}) = \sum_{r,p,q=1}^{2S+1} c_{rpq} (S_n^x)^r (S_n^y)^p (S_n^z)^q \quad (\text{B17})$$

then we have the equation

$$\begin{pmatrix} 0 & ih_z & -ih_y \\ -ih_z & 0 & ih_x \\ ih_y & -ih_x & 0 \end{pmatrix} \begin{pmatrix} \langle P_n S_n^x \rangle \\ \langle P_n S_n^y \rangle \\ \langle P_n S_n^z \rangle \end{pmatrix} = \frac{1}{2} \begin{pmatrix} \coth\left(\frac{\beta\tilde{\omega}}{2}\right) & -ih_z & ih_y \\ ih_z & \coth\left(\frac{\beta\tilde{\omega}}{2}\right) & -ih_x \\ -ih_y & ih_x & \coth\left(\frac{\beta\tilde{\omega}}{2}\right) \end{pmatrix} \begin{pmatrix} \langle [S_n^x, P_n] \rangle \\ \langle [S_n^y, P_n] \rangle \\ \langle [S_n^z, P_n] \rangle \end{pmatrix}$$

If we take this set of equations for $P_n \equiv P(\{S_n^\alpha\})$, along with the regularity conditions

$$(S_n^x)^2 + (S_n^y)^2 + (S_n^z)^2 = S(S+1) \quad (\text{B18})$$

$$[S_n^\alpha, S_n^\beta] = i\epsilon_{\alpha\beta\delta} S_n^\delta \quad (\text{B19})$$

$$\langle S_n^z \rangle h_x = \langle S_n^x \rangle h_z, \quad \langle S_n^x \rangle h_y = \langle S_n^y \rangle h_x, \quad \langle S_n^y \rangle h_z = \langle S_n^z \rangle h_y \quad (\text{B20})$$

and the usual spin algebra identities for spin- S degrees of freedom, we find that the system of equations for the two-point functions $\langle S_n^\beta S_n^\alpha \rangle$ and $\langle S_n^{x,y} \rangle$ can be solved as a function of $\langle S_n^z \rangle$, i.e., we need one extra equation to solve the system. This can be obtained from the identity:

$$\prod_{r=-S}^S (S_n^z - r) = 0 \quad (\text{B21})$$

which clearly becomes more and more complicated as the spin S is increased.

(ii) Specific Cases involving electronic and nuclear spins: As a first check, we can take $S = 1/2$. In that case we find that $(S_n^z)^2 = 1/4$, which provides the extra equation needed for the solution. The system of equations then results in:

$$\langle S_n^\mu \rangle = \frac{h_\mu}{2} \tanh\left(\frac{\beta\tilde{\omega}}{2}\right), \quad \langle (S_n^\mu)^2 \rangle = \frac{1}{4}, \quad \langle S_n^\mu S_n^\nu \rangle = i \frac{\epsilon_{\mu\nu\delta} h_\delta}{4} \tanh\left(\frac{\beta\tilde{\omega}}{2}\right) \quad (\text{B22})$$

which is the expected result from the RPA calculation for a spin 1/2.

Consider now the case of $S = 1$, for which the extra equation reads:

$$(S_n^z)^3 = S_n^z \quad (\text{B23})$$

Hence, we must obtain statistical averages for $(S_n^z)^3$ as well, by setting $P_n = (S_n^x)^p (S_n^y)^q (S_n^z)^r$ (for this case $p, q, r = 0, 1, 2$ is sufficient) and deal with a larger system of equations. We can then see how a general rule for arbitrary spins emerges - this was derived by Wang et al. [44] - and one finds:

$$\langle S_n^z \rangle = \frac{[(2S+1)R - Q_z](Q_z + R)^{2S+1} + [(2S+1)R + Q_z](Q_z - R)^{2S+1}}{2R^2 [(Q_z + R)^{2S+1} - (Q_z - R)^{2S+1}]} \quad (\text{B24})$$

where we have defined

$$R = 1/|h_z|, \quad Q_z = \frac{\coth\left(\frac{\beta\tilde{\omega}}{2}\right)}{h_z} \quad (\text{B25})$$

Now it is easy to see how to deal with a set of coupled nuclear and electronic spins. We first consider the nuclear spin averages. As an example, take the case where $I_n = 7/2$ (the case of the *Ho* nuclear spins in the *LiHoF* system). Then for the nuclear spin averages we get

$$\langle I_n^z \rangle = \frac{1}{2Q_z} + \frac{2Q_z}{Q_z^2 + R^2} + \frac{8Q_z(Q_z^2 + R^2)}{Q_z^4 + 6Q_z^2R^2 + R^4} \quad (\text{B26})$$

$$\langle I_n^y \rangle = \frac{h_y}{h_z} \langle I_n^z \rangle, \quad \langle I_n^x \rangle = \frac{h_x}{h_z} \langle I_n^z \rangle \quad (\text{B27})$$

As these equations depend on $\tilde{\omega} = \tilde{\omega}(\langle I_n^x \rangle, \langle I_n^y \rangle, \langle I_n^z \rangle)$, we have to solve them numerically.

If we now consider the full *LiHo* system, ie., with a coupling between an Ising electronic system ($S = 1/2$) and a spin bath ($I = 7/2$), the self-consistency equations are coupled for all values of the magnetization. We then find:

$$M_\mu = \frac{h_\mu^S}{2} \tanh\left(\frac{\beta\tilde{\omega}}{2}\right) \quad (\text{B28})$$

$$m_\mu = \frac{h_\mu^B}{2} \left[\tanh\left(\frac{\beta\tilde{\Omega}}{2}\right) + 2 \tanh(\beta\tilde{\Omega}) + 4 \tanh(2\beta\tilde{\Omega}) \right]$$

for the electronic system and nuclear bath magnetizations respectively, where $h_\mu^{S(B)} = H_\mu^{S(B)}/\tilde{\omega}(\tilde{\Omega})$ represent the system(environment) normalized fields respectively, and $\tilde{\omega}(\tilde{\Omega})$ represent the normalized eigenenergies of the system(environment) respectively. Note that now $h_\mu^S = h_\mu^S(M_\mu, m_\mu)$, $h_\mu^B = h_\mu^B(M_\mu)$, $\tilde{\Omega} = \tilde{\Omega}(M_\mu)$ and $\tilde{\omega} = \tilde{\omega}(M_\mu, m_\mu)$. We can easily obtain the $T = 0$ limit from these expressions; we get:

$$M_\mu = \frac{1}{2} \frac{B_\mu + M_\mu V_0^\mu - A_0^\mu m_\mu}{\sqrt{(B_x - A_0^x m_x)^2 + A_0^y m_y^2 + (V_0^z M_z - A_0^z m_z)^2}} \quad (\text{B29})$$

$$m_\mu = -\frac{7}{2} \frac{A_\mu M_\mu}{\sqrt{(A_0^x M_x)^2 + (A_0^y M_y)^2 + (A_0^z M_z)^2}} \quad (\text{B30})$$

Therefore, we can directly substitute m_μ into the system magnetization. Since we are interested in the behavior at large B_x , in order to see if the QPT can be blocked, we note that in the asymptotic limit $B_x \gg A_\mu, V_0^z$, one finds:

$$M_x \lesssim \frac{1}{2}, \quad M_z \simeq \frac{m_z V_0^z}{2B_x} + \frac{7(A_0^z)^2 m_z}{4B_x \sqrt{(A_0^x m_x)^2 + (A_0^z m_z)^2}} \quad (\text{B31})$$

Clearly in the second equation we could have $M_z = 0$ as a solution of the system, meaning that a QPT would exist. However, if we assume the highly anisotropic case $A_z \neq 0$, $A_x = 0$, we find:

$$M_z = \frac{1}{2} \frac{M_z V_0^z + \frac{7}{2} A_0^z}{\sqrt{B_x^2 + (V_0^z M_z + \frac{7}{2} A_0^z)^2}} \simeq \frac{M_z V_0^z}{2B_x} + \frac{7A_0^z}{4B_x} \quad (\text{B32})$$

which proves that M_z will always have a remnant magnetization blocking the QPT at $T = 0$ for all B_x (the second equation can never be fulfilled when $M_z = 0$). Hence, the longitudinal hyperfine coupling blocks the phase transition as one would expect.

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